

Multiphoton Processes in Driven Mesoscopic Systems.

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We study the statistics of multi-photon absorption/emission processes in a mesoscopic ring threaded by an harmonic time-dependent flux $\Phi(t)$. For this sake, we demonstrate a useful analogy between the Keldysh quantum kinetic equation for the electrons distribution function and a Continuous Time Random Walk in energy space with corrections due to interference effects. Studying the probability to absorb/emit n quanta $\hbar\omega$ per scattering event, we explore the crossover between ultra-quantum/low-intensity limit and quasi-classical/high-intensity regime, and the role of multiphoton processes in driving it.

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I. INTRODUCTION

Recently a surge of interest in the dynamical properties of mesoscopic/nanoscale electronic systems has motivated a number of theoretical and experimental studies on the physics of electronic devices subject to the driving of external fields. This main theme, pioneered in Ref. 1, embraces a number of interesting issues such as the study of the influence of microwave driving on transport through chaotic scatterers², the phenomenon of adiabatic quantum pumping³, as well as diffusion and localization^{4,5,6,7} in energy space in quantum chaotic systems/disordered quantum dots. In a broader context, the effect of the driving of external microwave fields has been shown to lead to an intriguing zero resistance state in quantum Hall systems⁸, and is currently studied as a tool to control the coherent dynamics of superconducting Josephson qubits⁹.

Investigations of periodically driven mesoscopic systems/quantum dots addressed mostly the limit of low intensity driving^{2,6,7}. In this case, electrons have enough time to explore ergodically all available phase space before performing a single photon assisted transition in energy space. This makes it possible to use an effective time dependent Random Matrix Theory to describe the dynamics of the system. On the other hand, as beautifully shown by recent experiments in superconducting qubits⁹ and in the quantum Hall regime⁸, as the intensity of driving increases one should expect both an enhancement of the probability of single-photon processes, and the emergence of multi-photon processes/resonances in the dynamical properties of the system under study.

The goal of this paper is to characterize the influence of multiphoton processes on the dynamical properties of mesoscopic electronic systems, concentrating on their effect on the electron dynamics in energy space (diffusion/localization). Diffusion and localization in energy space, as well as of multi-photon processes, have been the subject of a number of studies in the context of the optics

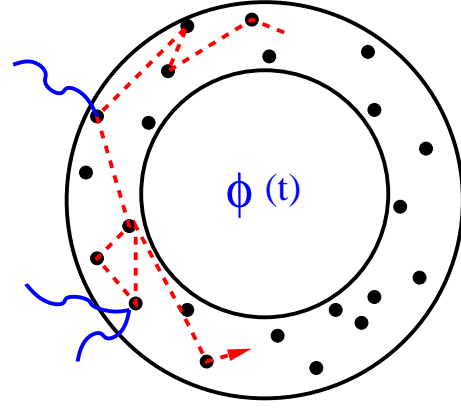


FIG. 1: The physical system under study, a diffusive quasi one dimensional ring threaded by a time dependent flux $\Phi(t)$. For an harmonic time dependence $\Phi(t) = \Phi \cos(\omega t)$, the scattering of electrons off impurities induces transitions in energy space quantized in units $\hbar\omega$. The statistics of such transitions and its physical consequences as the intensity of driving grows are described by Eq.(8) and Eq.(5).

of complex atoms/molecules¹⁰. In these systems the underlying electron dynamics is typically very complex and a statistical description, either equivalent to random matrix theory or explicitly using it, is compulsory. In contrast, in the present study we go beyond random matrix theory focusing on a model mesoscopic system, a diffusive quasi-one dimensional ring threaded by an oscillating flux (see Fig.1 and Eq.(14)), where the underlying microscopic dynamics can be studied in detail. We explore how multiphoton processes proliferate as the driving amplitude is increased or the driving frequency is decreased, study the resulting crossover from ultra-quantum/low-intensity limit to quasi-classical/high-intensity limit, and extract

its physical consequences. On the theoretical side, we demonstrate and use extensively an interesting analogy between the quantum kinetic equation for the electron distribution function and the recursion relation defining a Continuous Time Random Walk¹¹ in energy space.

The rest of the paper is organized as follows. In Sec. II we present qualitatively the results of our analysis of diffusion in energy space and of multiphoton processes based on the mapping of the problem onto a continuous time random walk in energy space. This mapping is derived in full detail in Sec. III using the Keldysh technique. Finally, in Sec. IV we present our conclusions.

II. QUALITATIVE ANALYSIS

In this section we start by summarizing the qualitative picture emerging from our analysis. The elementary time scale controlling the dynamics of energy absorption/emission is the mean free time τ . Indeed, in a diffusive quasi-1d ring threaded by a flux $\Phi(t) = \bar{\Phi} \cos(\omega t)$, energy changes quantized in units $\hbar\omega$ occur provided an electron scatters off an impurity. This is due to the fact that during the ballistic trajectory in between scattering events the flux perturbation $V(t) = -A(t) \hat{v}$, \hat{v} being the velocity operator, commutes with the unperturbed Hamiltonian $H_0 = m\hat{v}^2/2$, therefore causing no transitions whatsoever.

In the *ultra-quantum* limit of weak perturbations single-photon processes dominate. In other words, in one scattering event an electron may either absorb/emit one quantum, or scatter elastically. In particular, the probability P_Ω to make a transition of energy Ω in energy space in one scattering event is given by

$$P_\Omega = (1 - p)\delta(\Omega) + \frac{p}{2} [\delta(\Omega - \hbar\omega) + \delta(\Omega + \hbar\omega)], \quad (1)$$

where $p \propto (\bar{\Phi}/\Phi_0)^2 \ll 1$, Φ_0 being the flux quantum.

On the other hand, in the opposite limit of high intensities of the perturbation it is natural to expect *quasi-classical* continuous energy absorption described by a Drude-like picture. According to this picture an electron moving ballistically between two scattering events (at times t' and t , respectively) acquires an energy $\int_{t'}^t dt'' ev_F \vec{E}(t'') \cdot \hat{n}$, where $\vec{E}(t) = -\partial_t \vec{A}(t)$ is the electric field generated by the time dependent flux, and \hat{n} is the momentum direction in the d -dimensional space. Let us introduce the probability density $\mathcal{P}_\Omega(t, t')$ of changing the energy by Ω between two successive scattering events at t' and t . Given the Poisson distribution

$$\psi(t - t') = \frac{1}{\tau} e^{-|t-t'|/\tau}, \quad (2)$$

of time intervals $|t - t'|$, neglecting acceleration by an electric field $\vec{E}(t)$, and assuming isotropic scattering, one

may immediately write

$$\mathcal{P}_\Omega(t, t') = \psi(t - t') \left\langle \delta \left(\Omega - \int_{t'}^t dt'' ev_F \vec{E}(t'') \cdot \hat{n} \right) \right\rangle_{\hat{n}} \quad (3)$$

where $\langle * \rangle \equiv \int d\hat{n} (*)$ denotes the averaging over momentum directions. For an harmonic time dependence $E(t) = E_0 \cos(\omega t)$, and at low frequencies $\omega\tau \ll 1$, one obtains

$$P_\Omega = \int_{-\infty}^t \langle \mathcal{P}_\Omega(t, t') \rangle_T dt' = \begin{cases} \langle \frac{E_1(\frac{|\Omega|}{2\Omega_0 \sin(\omega\tau)})}{2\Omega_0 \sin(\omega\tau)} \rangle_T, & 3d \\ \langle \frac{K_0(\frac{|\Omega|}{\pi\Omega_0 \sin(\omega\tau)})}{\pi\Omega_0 \sin(\omega\tau)} \rangle_T, & 2d \end{cases} \quad (4)$$

where $\langle * \rangle_T = \int_{-\frac{\pi}{\omega}}^{\frac{\pi}{\omega}} \frac{dt\omega}{2\pi} (*)$ denotes averaging over the period, $E_1(z) = \int_1^\infty e^{-zt} \frac{dt}{t}$ is the exponential integral function, $K_0(z)$ is the Bessel function, and $\Omega_0 = eE_0 v_F \tau$.

Multiphoton processes drive the crossover between discrete energy absorption in the ultra-quantum limit [Eq.(1)], and continuous energy absorption in the quasi-classical limit [Eq.(4)]. As shown below, the crossover probability function P_Ω may be written as $P_\Omega = \sum_n P_n \delta(\Omega - \hbar n \omega)$. In the low frequency limit $\omega\tau \ll 1$, and for isotropic 3d scattering, we obtain

$$P_n = \mathcal{E}^{2n} A_n {}_3\mathcal{F}_2 [a_n, b_n, -16\mathcal{E}^2] \quad (5)$$

where $\mathcal{E} = eE_0 v_F \tau / (\hbar\omega)$, ${}_3\mathcal{F}_2$ is a generalized hypergeometric function, $A_n = (2^{2n} \Gamma[n + 1/2]) / (\sqrt{\pi} (1 + 2n) \Gamma[n + 1])$, $a_n = \{n + 1/2, n + 1/2, n + 1/2\}$, and $b_n = \{n + 3/2, 1 + 2n\}$. These functions are plotted for selected values of n in Fig.1.

At low intensities ($\mathcal{E} \ll 1$) the probability to absorb/emit n photons is

$$P_n = A_n \mathcal{E}^{2n} \quad \mathcal{E} \ll 1. \quad (6)$$

Therefore, multi-photon processes are exponentially suppressed. Neglecting them we obtain Eq.(1) with $p = \mathcal{E}^2/6$. As the intensity grows, higher order processes become increasingly probable at the expense of single (or in general low) order ones, as indicated by the fact that for $\mathcal{E} > 2$, $P_1(\mathcal{E})$ starts decreasing. At $\mathcal{E} \gg 1$ Eq.(5) can be approximated as follows:

$$P_n \propto \frac{1}{\mathcal{E}} \begin{cases} \ln^2(\mathcal{E}/n), & \mathcal{E} \gg n \\ \exp[-n/\mathcal{E}], & \mathcal{E} \ll n \end{cases} \quad (7)$$

Note that in the interval $1 \ll n \ll \mathcal{E}$ the probability of absorbing/emitting n photons decrease very slowly with increasing n which leads to a proliferation of multi-photon processes at large \mathcal{E} .

The two distinct regimes of rare ($\mathcal{E} \ll 1$) and proliferating ($\mathcal{E} \gg 1$) multiple photon processes have been discussed by Keldysh¹² in his seminal work on atom ionization. In this case the two regimes are classified by the ratio $\gamma^{-1} = \omega_t/\omega$ of the inverse time $\omega_t = eE_0/\sqrt{mI_0}$ of tunneling through the potential barrier titled by the electric field E_0 , $I_0 \gg \hbar\omega$ being the ionization threshold,

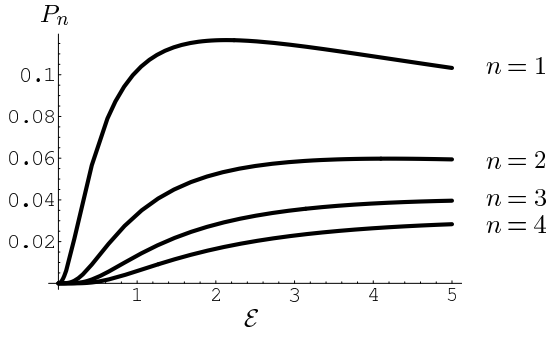


FIG. 2: The probability to absorb/emmit n photons in a scattering event P_n plotted as a function of the intensity parameter \mathcal{E} for selected values of n . At low intensities, single photon processes dominate. At higher intensities, higher order multiphoton processes become increasingly important, driving a quantum-to-classical crossover [see text].

and of the frequency of the applied electromagnetic field ω . The qualitative connection between Ref.[12] and our problem is obtained by identifying γ^{-1} with \mathcal{E} , and I_0 with $1/\tau$ in the present analysis.

Let us now make the qualitative considerations above more precise. As shown in Sec.III in terms of a Keldysh diagrammatic analysis^{7,13,14}, the dynamics of energy absorption/emission in the system at hand may be conveniently described as *random walk* in the energy space described by the recursion relation for the electron energy distribution function $f_t(E)$

$$f_t(E) = \int_0^t dt' \int_{-\infty}^{+\infty} d\Omega \mathcal{P}_\Omega(t, t') f_{t'}(E - \Omega). \quad (8)$$

Neglecting weak localization effects controlled by the parameter $\lambda_f/v_F\tau \ll 1$ and effects of dynamic localization controlled by the parameter $\delta/(v_F^2\tau A(t)^2) \ll 1$ (where δ is the mean separation of electron levels in a finite system)⁶, the kernel \mathcal{P}_Ω , is given by the product of two functions $\mathcal{P}_\Omega(t, t') = \psi(t - t')p_\Omega(t, t')$. The function $\psi(t - t')$, given by Eq.(2), is the distribution of the ballistic time of flight $|t - t'|$, which may be interpreted as the continuous waiting time in between steps of a random walk in the energy space. The other function, $p_\Omega(t, t')$, is the conditional probability to absorb/emmit an energy Ω during the ballistic flight and is given by $p_\Omega(t, t') = \int d\eta e^{-i\Omega\eta} \tilde{p}_\eta(t, t')$, where

$$\tilde{p}_\eta(t, t') = \left\langle e^{i v_F \hat{n} \cdot [\int_{t'+\eta/2}^{t+\eta/2} dt'' e \vec{A}(t'')] } \right\rangle_{\hat{n}}. \quad (9)$$

This result holds for a generic time dependence of $\vec{A}(t) = \hat{n}_x \Phi(t)/L$ as long as $e|\vec{A}(t)|\lambda_F \ll 1$. A random walk of the type defined by the recursion relation Eq.(8) is known in literature as a *Continuous Time Random Walk* (CTRW)¹¹.

One may now easily derive the crossover probability function Eq.(5). Indeed, in the case of harmonic flux

$A(t) = \frac{E_0}{\omega} \cos(\omega t)$, one obtains

$$p_\Omega(t, t') = \sum_{n=-\infty}^{+\infty} \delta(\Omega - n\hbar\omega) p_n(t, t') \quad (10)$$

$$p_n(t, t') = \left\langle J_{2n} \left[2 \frac{\mathcal{E}}{\omega\tau} \hat{n} \cdot \hat{n}_x [\cos(\omega t) - \cos(\omega t')] \right] \right\rangle_{\hat{n}},$$

where $J_{2n}(z)$ is a Bessel function and $\mathcal{E} = eE_0v_F\tau/\hbar\omega$. Though $p_n(t, t')$ does not explicitly depend on τ , the probability (averaged over the period T of flux oscillations)

$$P_n = \int_{-\infty}^0 dt' \psi(t - t') \langle p_n(t, t') \rangle_T, \quad (11)$$

of a multi-photon process between two successive scattering events does depend on the mean free time τ through the function $\psi(t)$. For $\omega\tau \ll 1$ one expands the difference of cosines in Eq.(10), introduce a new variable $(t - t')/\tau$ and immediately concludes that $P_n \equiv P_n(\mathcal{E})$ is a function of $\mathcal{E} = eE_0v_F\tau/(\hbar\omega)$. Performing explicit integrations in $3d$ leads finally to Eq.(5). Similar results may be derived in the high-frequency limit $\omega\tau \gg 1$. In this case where one may set $\psi(t - t') \approx 1$ and observe that $P_n \equiv P_n(\tilde{\mathcal{E}})$ is a function of the τ -independent parameter $\tilde{\mathcal{E}} = eE_0v_F/\hbar\omega^2$.

It is now possible to show directly that the discrete probability distribution P_n interpolates between *ultra-quantum* limit and *quasi-classical* continuous energy absorption. First of all, one may directly compare Eqs.(7) and (4): calculating the average over one period in Eq.(4) (which is dominated by small $t \ll \omega^{-1}$ for $\Omega \ll \Omega_0$ and by $\omega t \approx \pi/2$ for $\Omega \gg \Omega_0$) and replacing $\Omega \rightarrow n\hbar\omega$ one indeed obtain the classical result of Eq.(7). An alternative way to see the crossover, is to compute the moments of the number of absorbed/emitted photons $\langle (n)^m \rangle$ exactly using Eq.(5). One obtains

$$\begin{aligned} \langle (n)^2 \rangle &= \frac{1}{3} \mathcal{E}^2, \\ \langle (n)^4 \rangle &= \frac{1}{3} \mathcal{E}^2 + \frac{9}{5} \mathcal{E}^4, \\ \langle (n)^6 \rangle &= \frac{1}{3} \mathcal{E}^2 + 9 \mathcal{E}^4 + \frac{225}{7} \mathcal{E}^6, \\ \langle (n)^8 \rangle &= \frac{1}{3} \mathcal{E}^2 + \frac{189}{5} \mathcal{E}^4 + 450 \mathcal{E}^6 + 1225 \mathcal{E}^8. \end{aligned} \quad (12)$$

The *ultra-quantum* limit corresponds to all $\langle (n)^{2m} \rangle = \frac{1}{3} \mathcal{E}^2$, i.e. keeping only the first term in Eq.(12). On the other hand, the classical distribution [$3d$ -case in Eq.(4)] leads to moments $\langle (\Omega)^{2m} \rangle$ coinciding with the last terms in Eq.(12) upon the replacement $n \rightarrow \Omega/\hbar\omega$. This shows again that multi-photon processes drive the system at large intensities $\mathcal{E} \gg 1$ towards its quasi-classical limit.

The table of moments Eq.(12) can in principle be extracted from the smooth envelope of the distribution function $f_t^{env}(E)$, using the standard techniques of the theory of random walks¹¹ to translate the properties of

the product of a retarded and advanced Green's function appearing in Eq.(18) generates a diffusion propagator. More specifically, δG^K admits a diagrammatic representation in terms of a loose diffuson [see Fig.(4-(a))], which formally amounts to the equation

$$\delta G^K = 2\pi i\nu \int dt' dt'' \mathcal{D}_\eta(t, t') \mathcal{L}_\eta(t', t'') h_0(\eta), \quad (19)$$

where, D is the standard diffuson [see Fig.(4-(b))] solution of the equation

$$\begin{aligned} D^{-1} \otimes D_\eta &\equiv D_\eta(t, t') - \int dt'' \Pi_\eta(t, t'') D_\eta(t'', t') \\ &= \delta(t - t'). \end{aligned} \quad (20)$$

Neglecting interference effects, the kernel Π , as well as the vertex \mathcal{L} are given by

$$\begin{aligned} \Pi_\eta(t, t') &= \int d\eta' Tr \langle G^r(t_+, t'_+) \rangle \langle G^a(t'_-, t_-) \rangle / (2\pi\nu\tau) \\ \mathcal{L}_\eta(t, t') &= \int d\eta' Tr \langle G^r(t_+, t'_+) \rangle \langle G^a(t'_-, t_-) \rangle [V(t'_-) \\ &\quad - V(t'_+)] / (2\pi\nu i), \end{aligned}$$

where $t_\pm = t \pm \eta/2$ and $t'_\pm = t' \pm \eta'/2$, [see Fig.(4(b)-(c))]. The Tr symbol stands for the trace over the coordinate indices; in particular it implies $\int d\mathbf{p} = \int d\epsilon(p) \int d\hat{n}$ in the momentum representation where the disorder averaged Green's functions $\langle G^{r,a}(t, t'; \mathbf{p}) \rangle$ are diagonal. Preforming explicitly the trace, one obtains

$$\Pi_\eta(t, t') = \theta(t - t') \psi(t - t') p_\eta(t, t'), \quad (21)$$

$$\mathcal{L}_\eta(t, t') = \theta(t - t') \psi(t - t') \partial_{t'} p_\eta(t, t'), \quad (22)$$

where $\psi(t)$ is given by Eq.(2) and

$$\begin{aligned} p_\eta(t, t') &= \int d\hat{n} \exp \left[i v_F \hat{n} \cdot \hat{n}_x \left(\int_{t'+\eta/2}^{t+\eta/2} dt'' eA(t'') \right. \right. \\ &\quad \left. \left. - \int_{t'-\eta/2}^{t-\eta/2} dt'' eA(t'') \right) \right]. \end{aligned} \quad (23)$$

Finally we may express $h_t(\eta)$ in terms of \mathcal{D}, \mathcal{L} as

$$h_t(\eta) = \left(1 - \int dt' dt'' \mathcal{D}_\eta(t, t') \mathcal{L}_\eta(t', t'') \right) h_0(\eta). \quad (24)$$

Let us now show that the distribution function is in the kernel of the inverse diffusion propagator, i.e.

$$D^{-1} \otimes h_t(\eta) = 0. \quad (25)$$

First of all notice that

$$\begin{aligned} \tau \partial_{t'} \Pi_\eta(t, t') &= -\delta(t - t') + \Pi_\eta(t, t') + \mathcal{L}_\eta(t, t') \\ &= -[\mathcal{D}^{-1}]_\eta(t, t') + \mathcal{L}_\eta(t, t'). \end{aligned} \quad (26)$$

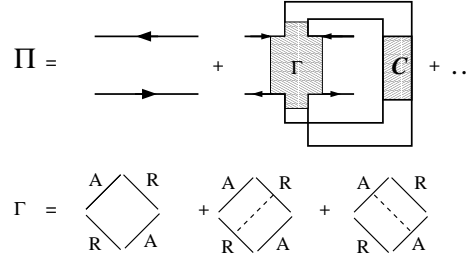


FIG. 5: Loop expansion for $P_\Omega(t, t')$, the insertion C being a cooperon⁷.

Now acting on Eq.(24) with the operator D^{-1} we obtain

$$\begin{aligned} [\mathcal{D}^{-1}] \otimes h_t(\eta) &= \left[\int dt' \mathcal{L}_\eta(t, t') - \tau \int dt' \partial_{t'} \Pi_\eta(t, t') \right. \\ &\quad \left. - \int dt' \mathcal{L}_\eta(t, t') \right] = 0. \end{aligned} \quad (27)$$

Notice at this point that Eq.(25) is equivalent to the recursion relation

$$h_t(\eta) = \int dt' \Pi_\eta(t, t') h_{t'}(\eta). \quad (28)$$

Since $\int dt' \Pi_{\eta=0}(t, t') = 1$, the latter may be equivalently stated as

$$\begin{aligned} f_t(\eta) &= \int dt' \Pi_\eta(t, t') f_{t'}(\eta) \\ &= \int_0^t dt' P_\eta(t, t') f_{t'}(\eta), \end{aligned} \quad (29)$$

where $P_\eta(t, t') = \psi(t - t') p_\eta(t, t')$. The Fourier Transform of this equation with respect to η gives the recursion relation defining the continuous time random walk, Eq.(8).

It is natural at this point to ask whether the formal description of the time evolution of the distribution function as an effective random walk may include interference/localization effects as well. Indeed, performing a one-loop analysis with accuracy \mathcal{E}^2 one may shown⁷ that the structure of Eq.(8) persists with a probability kernel schematically represented by the diagrams in Fig. (5). It is clear however that, in the presence of interference, the Markovian nature of the random walk is lost¹⁷. In particular, upon time integration we obtain a probability distribution P_Ω given by Eq.(1) with $p(t) \simeq \frac{\mathcal{E}^2}{3} \left(1 - \sqrt{\frac{t}{t^*}} \right)$, where the driving time t is the time since the turning on of the perturbation, $t^* = 2\pi^3 D_E / (\Omega^2 \delta^2)$ is the localization time in energy space⁶, and we neglected all corrections independent of t . This result amounts to the weak localization suppression of the absorption rate $W(t)/W_0 \simeq 1 - \sqrt{t/t^*}$ due to weak dynamical localization⁶.

IV. CONCLUSIONS

In conclusion, we have studied the problem of energy absorption/emission in a mesoscopic ring threaded by an oscillating flux, focusing on the influence of multiphoton processes, and on the multiphoton driven a crossover from ultra-quantum/low-intensity limit, and quasi-classical/high intensity regime. We have shown that the dynamics of the distribution function may be mapped onto a continuous time random walk in energy space. Though in the present paper we focused on the

effect of a classical driving, recent advances in the field of circuit QED¹⁸ strongly suggest the possibility to investigate the role of single and multi-photon processes in the case of quantum driving, an interesting problem that remains a challenge for future work.

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